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Purcell effect investigation in organic Tamm plasmon structures

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Abstract. We provide a theoretical and experimental investigation of a Purcell enhancement in a Tamm-plasmon based microcavity structure with an organic (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP)) active layer. The microcavity structure was fabricated using several techniques as a magnetron deposition (Ta₂O₅/SiO₂ dielectric Bragg mirror) and a thermal evaporation (CBP organic layer and silver layer on the top). Dependency of the modal Purcell factor on emission angle and frequency was calculated using S-quantization formalism. The emission spectra and the rate of fluorescence intensity decay of the Tamm structure was experimentally measured in ultraviolet region. We show, that at the frequencies corresponded to the Tamm plasmon resonance, the rate of the CBP molecule fluorescence decay increasing. Maximum registered value of the Purcell factor is close to 3.

1. Introduction

A Tamm plasmon state is the novel electromagnetic field eigenstate, that localized on the edge between a metal layer and a distributed Bragg reflector was recently predicted theoretically [1,2] and registered experimentally [3]. Plenty of interesting phenomena like a single-photon emitting [4] and laser generation [5] was demonstrated in the Tamm plasmon-based structures. As well as increasing of the spontaneous emission rate [6]. Organic material based photonic devices has a bunch of applications today, and investigations in that area are developing actively. However, efficiency of the organic light emitting systems is limited by the transfer of carriers to a triplet states, radiative transition from which to the singlet ground state is forbidden. In that case, it is very interesting to apply features from plasmonics to the field of organic emitters to find ways to improve efficiency of existing organic light emitting systems.

In the current paper, we have investigated an influence of the Tamm plasmon state on spontaneous emission rate of the organic emitting material CBP (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl).



2. Experimental

Firstly, to fabricate the Tamm-plasmon based microcavity structure, a 5 pair distributed Bragg reflector (DBR) was evaporated on the SiO_2 substrate using magnetron deposition method. Thicknesses of the layers was chosen as 43.2 nm for Ta_2O_5 layer and 66.4 nm for SiO_2 layer to match with the Bragg condition. The photonic band gap (PBG) of fabricated Bragg reflector is centered at energy 3.17 eV, which corresponds to CBP emission region. Further, using Kurt. J. Lesker Spectros (organic and metal thin film deposition system consisting of a vacuum down to 1×10^{-8} mbar chamber) 26 nm CBP and 50 nm silver layers were deposited with evaporation rates 0.1 nm/s and 0.05 nm/s, respectively.

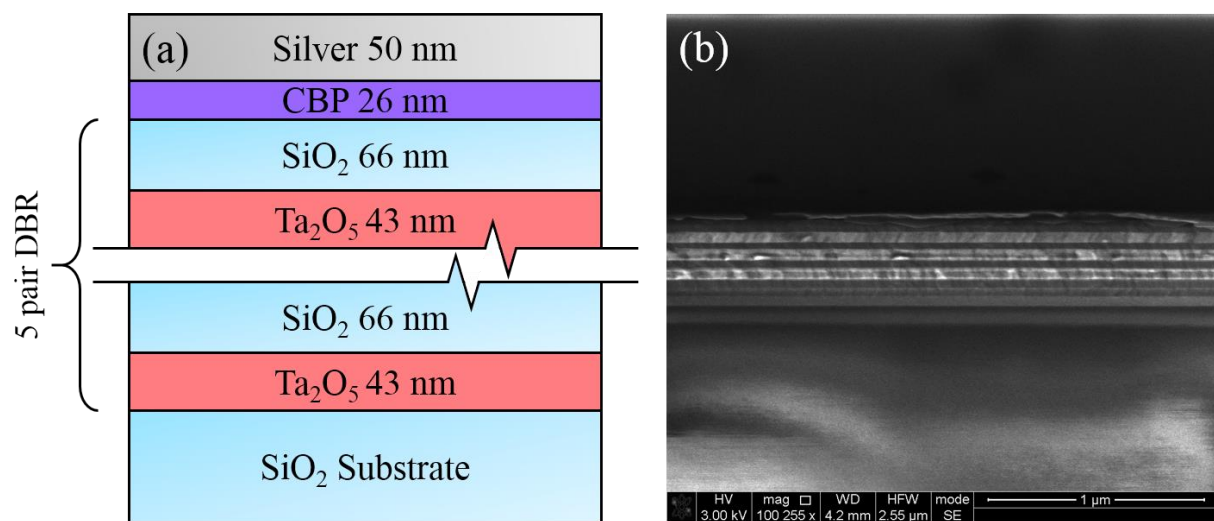


Figure 1(a, b). (color online) (a) A scheme of the Tamm plasmon microcavity structure. (b) Image of the structures fragment got by scanning electron microscopy.

Figure 1 (a) shows the scheme of the considered structure and (b) image of structures fragment got by scanning electron microscopy (SEM). Interfaces between DBR's top three $\text{Ta}_2\text{O}_5/\text{SiO}_2$ layers (figure 1 (b)) are very smooth, then the top two (CBP and silver) layers are not. It could be explained by the fact, that it is very hard to get a uniform fragment from a planar structure that included metal and organic layers. Also, a reference sample was fabricated: 26 nm CBP layer of the top of the sapphire substrate.

A fluorescence spectrum of CBP at room temperature (figure 2(b)) was measured by Jobin-Yvon Horiba Fluorolog FL3-22 spectrometer. Decay dynamics of CBP molecule's excited states coupled with the Tamm plasmon state and in a free space (reference sample) was measured by using a Time Correlated Single Photon Counting (TCSPC) scheme at room temperature. The structure was excited through the silver layer at 262 nm wavelength, the third harmonic of the output from a Coherent Mira 900F Ti:Sapphire oscillator tuned to a 786 nm central wavelength, with a 76 MHz repetition rate. An emission from the sample was collected at 45 degrees from the normal and at various energies (3.44 eV, 3.35 eV, 3.3 eV) in the CBP emission area.

3. Results and discussion

Theoretically, using S-quantization formalism [7] we analyzed properties and choose an optimal design of the Tamm structure. S-quantization method allow to calculate a modal Purcell factor – ratio between the spontaneous decay rate of the emitter placed in the cavity and the spontaneous emission rate for the emitter in the free space (homogeneous media). Obviously, to register effects of coupling

between the Tamm plasmon state and the organic active layer, frequency of the Tamm plasmon needed to be in a region where the CBP material emits effectively (near UV region).

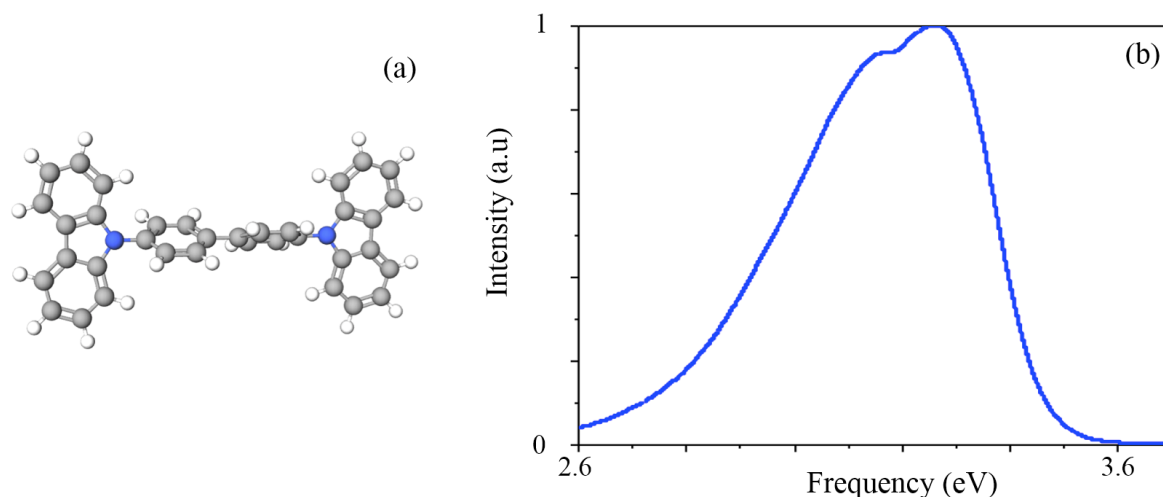


Figure 2(a, b). (color online) **(a)** A molecular structure of the CBP material (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl). **(b)** An emission spectrum of 26 nm CBP on sapphire reference sample.

A calculated distribution of the modal Purcell factor shown in figure 3(b). It can be seen, area that corresponds to the maximum of the calculated Purcell factor corresponds to the CBP emission area. Results of the TCSPC measurements are presented in figure 4 (a). Black line representing the decay curve of 26 nm CBP reference sample at the frequency 3.35 eV (a lifetime of the CBP singlet state is the same at energies 3.3 eV and 3.44 eV). Blue, green and red curves showing results of Tamm plasmon structure measurements at the frequencies 3.3 eV, 3.35 eV and 3.44 eV, respectively. It can be seen, that in comparison to the CBP reference sample emission in the free space, coupling with the Tamm plasmon mode increasing the decay rate (decreases the lifetime of the excited state) of the CBP emitter i.e. these results are confirmed the Purcell phenomenon in the considered structure. Thus, the experimental value of the Purcell factor could be estimated as the ratio between the lifetime of the excited state in case of the Tamm structure to the lifetime in case of the free space. Red curve (3.44 eV) is demonstrated the most significant decreasing of the CBP lifetime and corresponding to the Purcell coefficient 3.2. To compare experimental results with theoretically calculated, the modal Purcell factor distribution (figure 3(b)) needed to be integrate over all angles, to get the Purcell effect frequency spectrum in traditional meaning. In error limits, value of the experimental Purcell coefficient has good agreement with the theoretically estimated, but a bit higher. It could be explained by the non-radiative recombination that needed to take into account.

4. Conclusions

Theoretical and experimental investigation of the Purcell enhancement in the Tamm-plasmon based microcavity structure with the organic material (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP)) active layer was provided. Dependency of the modal Purcell factor on the emission angle and frequency was calculated using S-quantization formalism. The emission spectra and rate of fluorescence intensity decay of the Tamm structure was experimentally measured. Was shown, that at the frequencies corresponded to the Tamm plasmon resonance, the rate of the CBP molecule fluorescence decay increasing. A maximum registered value of the Purcell factor is close to 3.

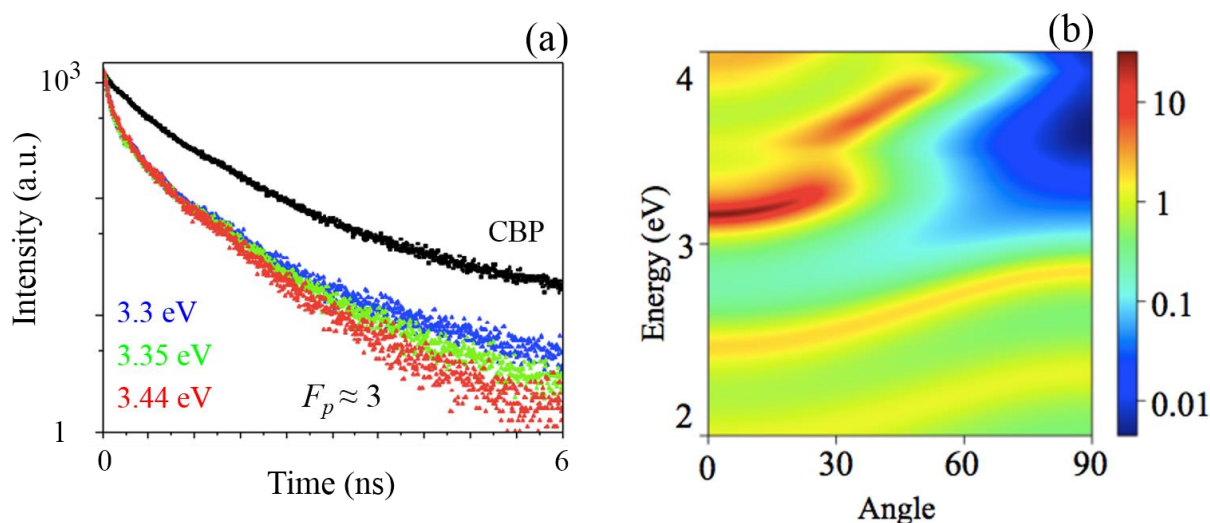


Figure 3(a, b). (color online) **(a)** Results of the TCSPC measurements. Black curve is decay curve of the 26 nm CBP reference sample at the frequency 3.35 eV. Blue, green and red curves are representing decay curves of Tamm plasmon structures at frequencies 3.3 eV, 3.35 eV, 3.44 eV, respectively. **(b)** The calculated distribution of the modal Purcell factor.

Acknowledgments

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